THERMAL EXPANSION AND MAGNETOSTRICTION STUDIES OF A KONDO LATTICE COMPOUND: $CeAgSb_2$

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We have investigated a single crystal of $CeAgSb_2$ using low field ac-susceptibility, thermal expansion and magnetostriction measurements. The thermal expansion coefficient α, exhibits highly anisotropic behaviour between 3K and 80K: α (for ΔL/L) $\perp$c exhibits a sharp peak at T_N followed by a broad maximum at 20K, while a sharp negative peak at T_N followed by a minimum at 20K has been observed for (ΔL/L $\|$) the c direction. The observed maximum and minimum in α(T) at 20K have been attributed to the crystalline field effect (CEF). The magnetostriction (MS) also exhibits anisotropic behaviour with a large MS along the c-axis.

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Recent studies on $RAgSb_2$ (R=rare earth) compounds have shown that these compounds crystallize in the tetragonal $ZrCuSi_2$ type structure [1-6]. Among these compounds, $CeAgSb_2$ is the most interesting. The resistivity and thermoelectric power of $CeAgSb_2$ show a typical Kondo lattice behaviour [1]. The magnetization exhibits strongly anisotropic behaviour with the easy c-axis of magnetization in the magnetic ordered state (probably a complex antiferromagnetic (AFM) state with a FM component) below 10K, while in the paramagnetic state there is an easy ab-plane [1,5]. The magnetization isotherm at 2K shows that the easy magnetization direction changes from the c-axis to the ab-plane above 1T field with the saturated moment μ^{sat}=1.1μ_B/Ce-ion for B$\perp$c and 0.37μ_B/Ce-ion for B$\|$c at 5.5T field. The neutron diffraction measurements at 2K show the presence of only a single magnetic Bragg peak, (1 0 1), with moment of 0.33μ_B/Ce-ion [5]. The zero-field μSR study shows well-defined frequency oscillations

with an anomalously low internal field of 53mT at the muon site, which is in agreement with the extremely low frequency (0.25MG) observed in a Shubnikov-de Haas study [4, 6].

In the present work we have investigated a single crystal of $CeAgSb_2$ using ac-susceptibility (χ_{ac}), thermal expansion, and magnetostriction measurements with the aim of throwing more light on the complex electronic and magnetic ground state. The single crystal of $CeAgSb_2$ was grown in an evacuated BN-crucible at 1350^{o}C by the Bridgemann method. The inductive component, χ'_{ac}(T) of the ac-susceptibility of $CeAgSb_2$ single crystal with B_{ac}(5G)$\|$c, exhibits a sharp peak at 9.7K which is due to the magnetic ordering of the Ce-moments (inset of Fig.1). χ'_{ac}(T) for $B_{ac}\perp$c also exhibits a similar peak, but the peak height is only 27% of $B_{ac}\|$c. The c-axis is therefore the easy magnetization direction at low temperature, which is in agreement with the dc magnetization study [4]. It would be interesting to compare the χ'_{ac}(T) signal of $CeAgSb_2$ with that from the ferromagnetic CePdSb [7]. Well below T_N (or T_C) χ'_{ac}(T) of $CeAgSb_2$ is very small, while that of CePdSb retains about 92% of its peak value. This again implies that the magnetic ground state of $CeAgSb_2$ is more complicated than that of a simple AFM or FM.

Fig.1 shows the linear thermal expansion (TE=ΔL/L) as a function of temperature for $CeAgSb_2$ single crystal parallel to c-axis (TE$\|$c) and perpendicular to c-axis (TE$\perp$c) along with the isostructural nonmagnetic reference polycrystalline $LaAgSb_2$. ΔL/L of $LaAgSb_2$ exhibits a typical behaviour expected for the thermally excited phonons. On the other hand, ΔL/L of $CeAgSb_2$ shows highly anisotropic behaviour, positive for TE$\perp$c and negative for TE$\|$c, with a sudden change at T_N in both the directions. The magnetic contribution to the thermal expansion coefficient, α_M(T) of $CeAgSb_2$ along both the directions was estimated by subtracting α(T) of $LaAgSb_2$. α_M(T) exhibits a sharp peak and a broad peak at T_N and 20K, positive for TE$\perp$c and negative for TE$\|$c, respectively. It should be noted that the α_M(T) of polycrystalline $CeAgSb_2$ also shows a broad peak at 18K, but no clear peak at T_N [3]. The absence of the peak at T_N in the polycrystalline sample might be due to the cancellation of positive (for TE$\perp$c) and negative (for TE$\|$c) contributions observed in the single crystal. The sharp peak at T_N in $CeAgSb_2$ single crystal arises due to the development of anisotropic spin-spin correlations because of the magnetic ordering of Ce-moments. On the other hand the broad peak (maximum and minimum) at 20K in both the directions has been attributed to the CEF effect on the J=5/2 state of the Ce^{3+} ion. This is consistent with our recent high resolution inelastic neutron scattering measurements on $CeAgSb_2$, which show two well defined crystal field excitations, at 5.1meV and 12.4meV, as expected for the tetragonal point symmetry of the Ce ion [8]. It is interesting

to note that the observed anisotropic behaviour of α_M(T) of $CeAgSb_2$ is very similar to that observed for $CeRhIn_5$ single crystal, which also has the tetragonal crystal structure [9]. The calculated α_M(T) for $CeRhIn_5$ on the basis of the CEF model exhibits a maximum and minimum around 25K for [100] and [001] directions, respectively [9]. In order to investigate the effect of magnetic field on the α_M(T) of $CeAgSb_2$, we have measured α_M(T) in an applied magnetic field of 8T (Fig.2). The observed sharp peak at T_N in zero field was almost suppressed in 8T field for both the directions.

We estimated the value of dT_N/dP=-0.088 (K/kbar), using the Ehrenfest relation and the heat capacity data from Ref.[2], which is in good agreement with the experimentally measured value of -0.095 (K/kbar) on the polycrystalline $CeAgSb_2$ [3]. The negative sign of dT_N/dP indicates that $CeAgSb_2$ is on the right-hand side of the Doniach phase diagram [3].

Fig.3 shows the magnetostriction (MS) isotherms measured at various temperatures for $(\Delta L/L)\|c$ and $(\Delta L/L)\perp c$ with applied fields $B\|c$ and $B\perp c$ directions. MS exhibits highly anisotropic behaviour with the largest length change for $(\Delta L/L)\|c$. Between 14Kand 20K MS exhibits a quadratic behaviour, for all measured directions (Figs.3a-d), taht could be understood on the basis of the free energy of the system in an applied field. An interesting behaviour of MS is observed for $(\Delta L/L)\perp c$ and $B\perp c$ geometry at low temperatures (Fig.3d). At 3K MS exhibits a peak at 3.3T, which is consistent with the observed peak in the magnetoresistance measurements and has been attributed to the field induced transition to the easy ab-plane of magnetization [4]. Furthermore, with increasing temperature from 3K, the position of the peak moves to a lower field, which indicates that the smaller critical field for the field induce transition. A small hysteresis in MS was observed at 3K suggesting the presence of a FM component. This result along with the absence of the domain walls contribution at low fields in MS of $CeAgSb_2$ indicates that the magnetic ground state is not a simple FM, but a complex AFM.

In conclusion, thermal expansion and magnetostriction measurements of $CeAgSb_2$ single crystal exhibit highly anisotropic behaviour. These results indicate that the anisotropic magnetic exchange and CEF-anisotropy are playing an important role.

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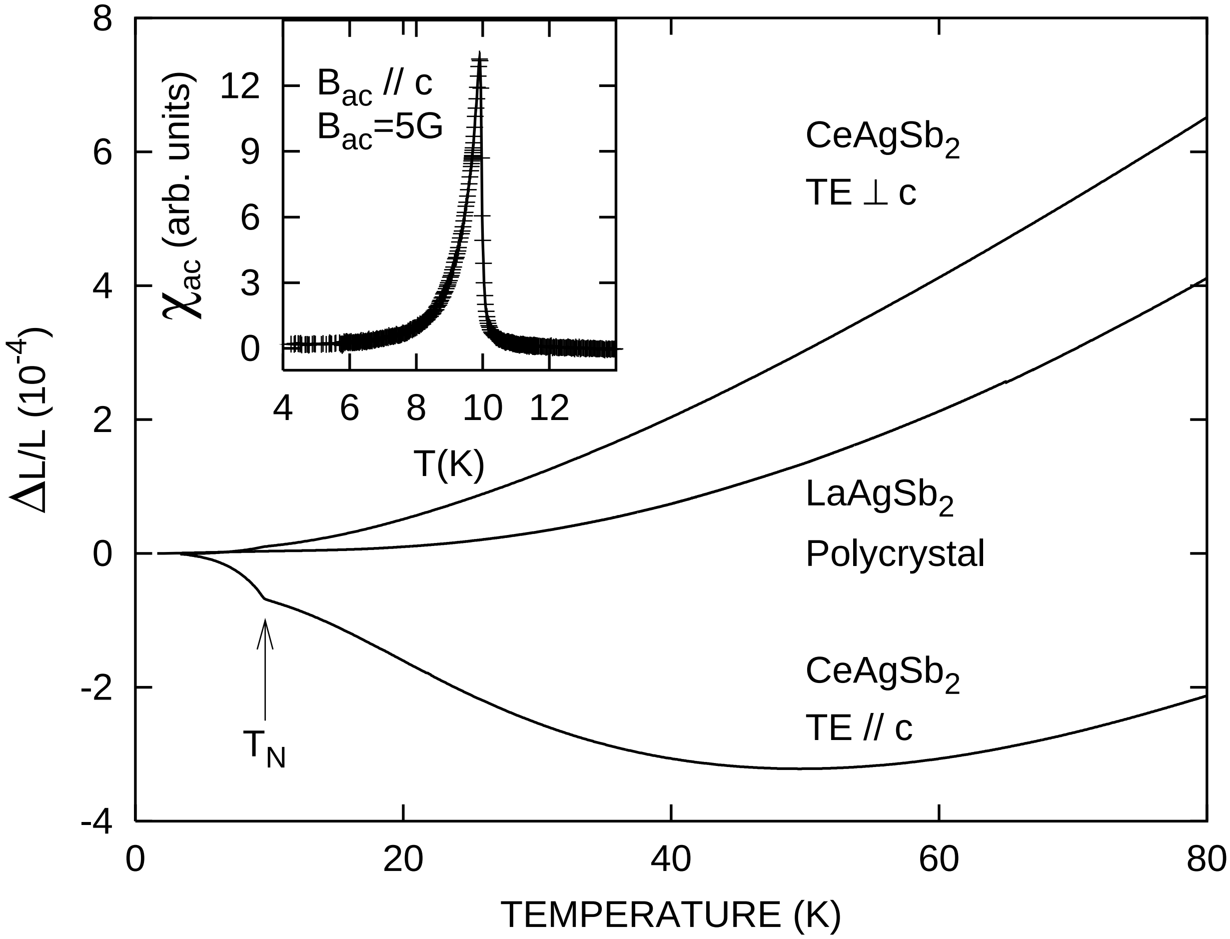
Figure captions

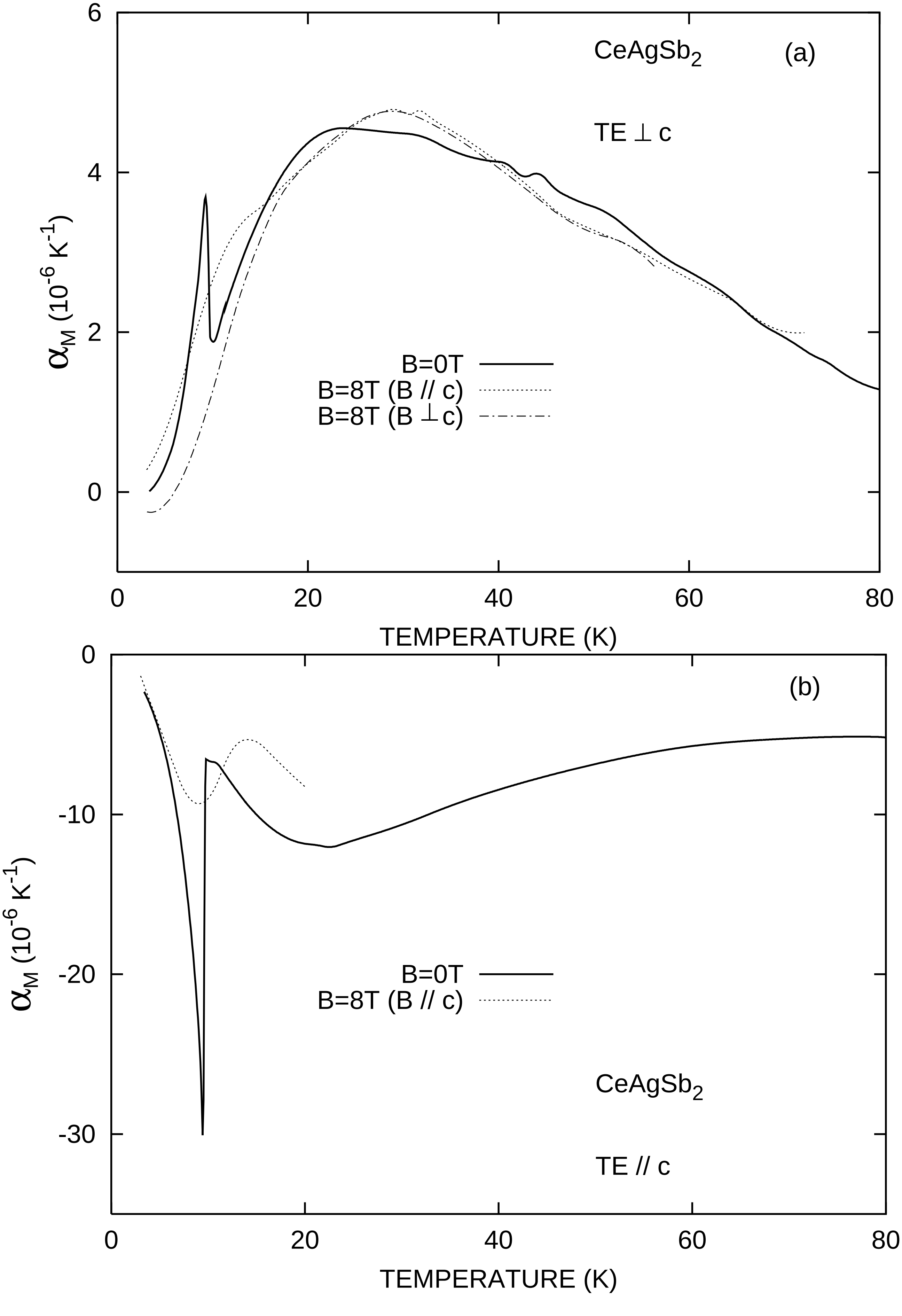
Fig 1. Linear thermal expansion (ΔL/L) versus temperature of $CeAgSb_2$ single crystal and $LaAgSb_2$ polycrystal. The inset shows temperature dependence of inductive, χ'_{ac}(T) component of the ac-susceptibility for $B_{ac}\|$c.

Fig.2 The magnetic contribution to the linear thermal expansion coefficient, α_M(T) of $CeAgSb_2$ single crystal in zero field and 8T field, (a) TE$\perp$c and, (b) TE$\|$c.

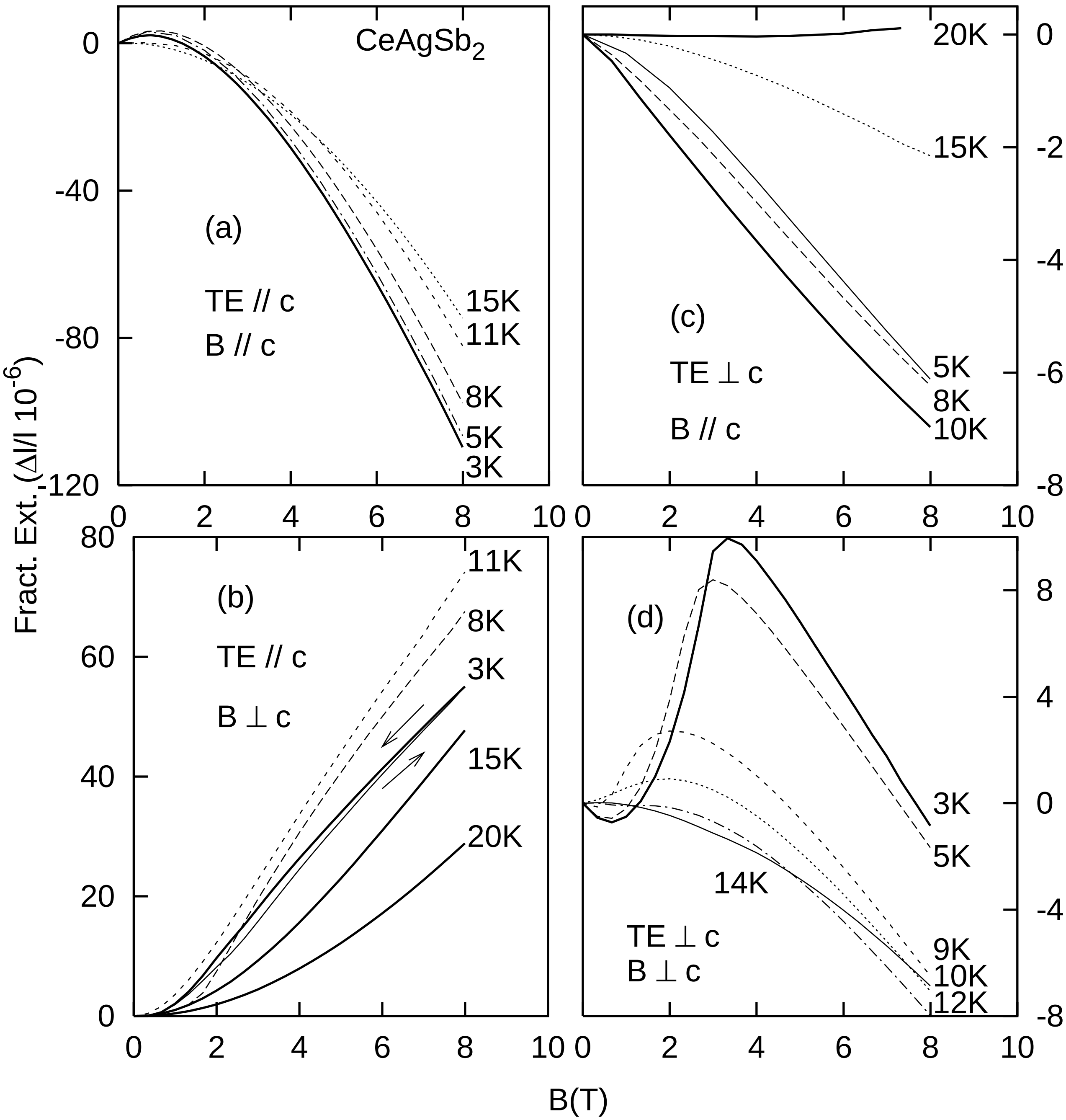
Fig.3 The magnetostriction isotherms at various temperatures of $CeAgSb_2$ single crystal, (a) and (b) (ΔL/L)$\|$c and, (c) and (d) (ΔL/L)$\perp$c, for both B $\|$c and B$\perp$c.



8
6
4
2
0
-2
-4
$\Delta L/L\ (10^{-4})$
0
20
40
60
80
TEMPERATURE (K)
$CeAgSb_2$
TE ⊥ c
$LaAgSb_2$
Polycrystal
$CeAgSb_2$
TE // c
T_N
B_{ac} // c
B_{ac}=5G
χ_{ac} (arb. units)
12
9
6
3
0
4
6
8
10
12
T(K)



CeAgSb2
(a)
TE ⊥ c
B=0T
B=8T (B // c)
B=8T (B ⊥ c)
αM (10-6 K-1)
TEMPERATURE (K)
0
20
40
60
80
6
4
2
0
(b)
B=0T
B=8T (B // c)
CeAgSb2
TE // c
αM (10-6 K-1)
TEMPERATURE (K)
0
-10
-20
-30



CeAgSb$_2$
(a)
TE // c
B // c
15K
11K
8K
5K
3K
(c)
TE ⊥ c
B // c
20K
15K
5K
8K
10K
(b)
TE // c
B ⊥ c
11K
8K
3K
15K
20K
(d)
TE ⊥ c
B ⊥ c
3K
5K
14K
9K
10K
12K
Fract. Ext. (Δl/l 10^{-6})
B(T)